Two-dimensional magnetic order of Er in $ErBa_2Cu_3O_7$

J. W. Lynn, T. W. Clinton, and W-H. Li
Center for Superconductivity Research, Department of Physics, University of Maryland, College Park, Maryland 20742 and National Institute of Standards and Technology, Gaithersburg, Maryland 20899

R. W. Erwin
National Institute of Standards and Technology, Gaithersburg, Maryland 20899

J. Z. Liu[a)]
Argonne National Laboratory, Argonne, Illinois 60439

R. N. Shelton and P. Klavins
Department of Physics, University of California, Davis, California 95616

Neutron diffraction has been used to study the magnetic order of the Er ions in superconducting $ErBa_2Cu_3O_7$. Above the three-dimensional (3D) Néel temperature ($T_N = 0.618$ K) a rod of scattering characteristic of two-dimensional (2D) behavior is unambiguously observed, showing that the magnetic interactions of the rare earth ions are highly anisotropic, while there are no significant correlations observed between the sheets of Er spins. The system orders two dimensionally, and as a necessary consequence 3D order also sets in at the same temperature. The order parameter is found to obey the exact Onsager solution for a $S = \frac{1}{2}$, 2D Ising antiferromagnet. At low T, two separate types of simple 3D antiferromagnetic structures are found, one characterized by a wave vector of $(\frac{1}{2}, 0,0)$, and the other by $(\frac{1}{2},0,\frac{1}{2})$.

Shortly after the discovery of superconductivity in the $YBa_2Cu_3O_7$ material, it was found that the substitution of the heavy rare-earth elements $\mathscr{R}$ for Y had little effect on the superconductivity in this system.[1-3] This observation, along with the low magnetic ordering temperatures for the rare earths,[4,5] indicates that the rare-earth ions are electronically isolated from the superconducting Cu-O planes, as well as from each other, and makes these materials ideal candidates to investigate the interplay between magnetism and superconductivity via the electromagnetic interaction.[6] The oxide superconductors are of course also very interesting because of the magnetism displayed by the Cu ions, but this falls into a different arena than the present study.[3]

The nature of the magnetic ordering of the rare earth ions in the 1-2-3 system has been something of a puzzle until recently.[7] The first neutron experiments were carried out on polycrystalline $ErBa_2Cu_3O_7$,[8] and showed a magnetic phase transition at $T_N \sim 0.6$ K which agreed with specific heat data.[4,5] The Er moments exhibited a two-dimensional character in the a-b plane with an ordering wave vector of $(\frac{1}{2}, 0)$, while there were no significant magnetic correlations which could be detected along the c axis down to the lowest temperatures that could be achieved at that time (0.33 K). This anisotropic behavior was expected since the interactions are primarily dipolar in origin, and the spacing between Er ions is three times larger along the c axis than in the a-b directions. Our present results on large high-quality single crystals directly reveal the existence of rods of scattering in reciprocal space, unambiguously demonstrating the expected 2D behavior. These results nullify the picture recently presented by Paul *et al.*,[9] where they were unable to observe this scattering. We believe that the anisotropy of the rare earth interactions is a general characteristic of the 1-2-3 class of materials, and we note that 2D behavior has also been recently observed in the 2-4-8 materials as well.[10]

In this paper we present and discuss our measurements both on our original powder sample of $ErBa_2Cu_3O_7$, measured to lower temperatures, and on high-quality single-crystal samples. Above the magnetic ordering temperature we observe a rod of scattering along $(\frac{1}{2},0,l)$, demonstrating the 2D behavior, while below the Néel temperature the 3D order parameter follows the Onsager solution for the $S = \frac{1}{2}$ 2D Ising model, analogous to the behavior observed for the prototype 2D Ising system K_2CoF_4.[11] There has recently been considerable confusion concerning the magnetic ordering of the rare-earth ions in these systems, and on the relationship between the 2D ordering and the 3D Bragg peaks observed in neutron scattering experiments. Thus, we will discuss in some detail the relationship between the 2D and 3D behavior, and also between measurements taken on powders versus single crystals.

The neutron experiments were conducted at the research reactor at the National Institute of Standards and Technology. The diffraction data were taken with a wavelength of 2.3509 Å and a pyrolytic graphite monochromator and filter. The properties of the polycrystalline sample have been described in our earlier report.[7] The growth technique for the single crystals yields (twinned) crystals which are uniformly and fully oxygenated during the growth process, eliminating the requirement of a post-oxygen anneal to make the samples superconducting.[12] The measured supercon-

[a)] Present address: Department of Physics, University of California-Davis, Davis, CA 95616.

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ducting transition temperature for the crystals used in the present measurements was 92.8 K, with a width of less than 1 K. Most of the neutron data were taken on a crystal which weighed 31 mg.

To understand the observations on this system, consider first the idealized situation where the interactions along the *c*-axis direction are identically zero, so that the rare-earth ions in the *a-b* planes (with interaction J_{ab}) are truly 2D in nature. Then, as the system is cooled we expect correlations to develop between the spins as kT becomes comparable to J_{ab}. In the case of an Ising system, where there is a unique direction in which the spins like to point, then true long range order will develop at finite temperature. The scattering intensity associated with this order will be given by

$$I_M(\mathbf{Q}) \propto \Big| \sum_{i,j}^{N} \langle S_{i,j} \rangle e^{2\pi i(hx_i + kyi)} \Big|^2,$$

where x and y are the spatial coordinates of the ions in the plane, $\mathbf{Q} = (h,k,l)$, and h, k, l are Miller's indices, and the sum is over all ions in the plane. Generally the moments are in a simple $\pm$ configuration, and this will determine the values of h and k where the Bragg scattering will occur. The essential point is that there is no restriction on the component l of momentum transfer perpendicular to the planes, and hence the Bragg scattering will occur along lines (rods) in reciprocal space rather than at points as in the 3D case. Figure 1 shows reciprocal space where most of our measurements were taken, and where the rods occur for the present case of interest.

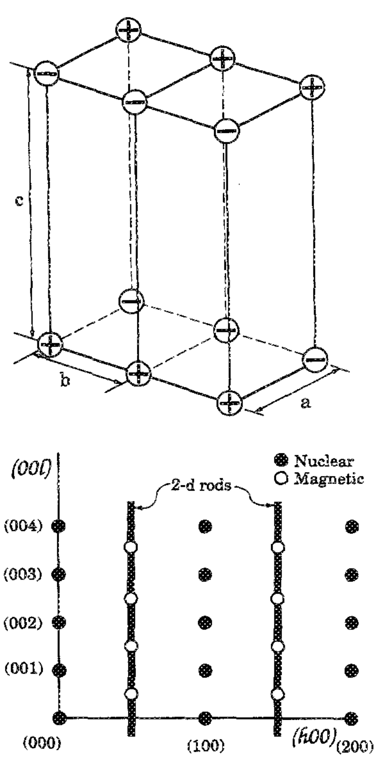

FIG. 1. Scattering plane in reciprocal space where most of our measurements were taken. The rods indicate where the 2D scattering is located, with the open circles signifying the positions of the 3D magnetic peaks. The nuclear reflections are indicated by the solid circles. The spin configuration is also shown.

Now assume that the planes are ordered two dimensionally, and introduce a small interaction J' between planes, with $|J'| \ll |J_{ab}|$, and consider the coupling between two planes. For an Ising system the nearest neighbors between adjacent planes are either parallel or antiparallel. However, if the first pair is parallel, then every pair is parallel since the spins are already ordered in the planes, while if the first pair is antiparallel then every pair is antiparallel. Hence, the two possible configurations will differ by an energy $\pm J' \langle S \rangle^2 (L_D)^2$, where L_D^2 is the average area of a domain within the layers. The interaction J' is thus greatly amplified because the planes are already ordered two dimensionally, and the energetics will then drive the system to order in the third dimension as well. This is the basic argument to explain why two-dimensional magnets exhibit three-dimensional Bragg peaks. The dynamics will display 2D behavior, and the order parameter will be controlled by the 2D behavior of $\langle S \rangle$, but the scattering will occur at 3D Bragg points as shown in Fig. 1, as has been found in all the "prototype" 2D magnets.[13]

Figure 2 shows the scattering we observed just above the ordering temperature, where the 3D Bragg peaks have not developed yet. Scans through the rod position show that the scattering peaks as expected for a 2D system. Also shown is the scattering intensity observed along the rod, and we see no significant correlations even though we are just above the 3D ordering temperature; the 3D Bragg peaks occur at $Q_z = 0.5$ and 1.5 in this plot. The gentle decrease with increasing Q_z is due to the form factor variation, and to a variation in the instrumental resolution. The observation of this uniform ridge of scattering demonstrates that the magnetic interactions within the planes are much stronger than between planes.

In the single crystals we have studied, we have not found any peaks with integer l, and hence we assume that the appropriate modulation vector for the ground state spin configuration is $(\frac{1}{2},0,\frac{1}{2})$. The spin structure for the system is

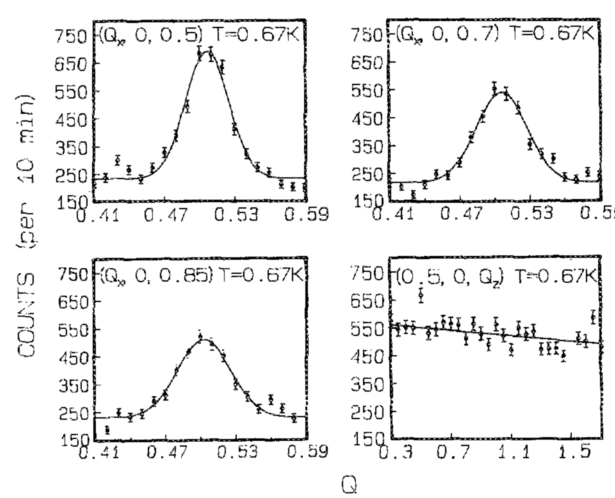

FIG. 2. Three scans across the "rod" of scattering in reciprocal space, showing the 2D character just above $T_N = 0.618$ K. The fourth scan shows that the scattering intensity does not vary significantly along the rod, indicating that there are no significant spin correlations between planes.

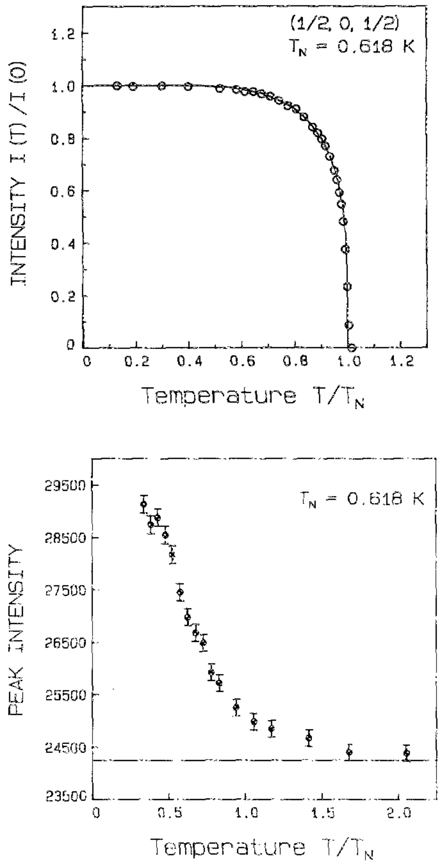

FIG. 3. The magnetic intensity vs temperature. The left part of the figure shows the results on a single crystal, and the solid curve is the Onsager result for a 2D Ising system. The right-hand side shows the data obtained on a powder.

shown in Fig. 1, and consists of ferromagnetic chains of spins along the b direction, with adjacent chains in the a-b plane coupled antiferromagnetically. In the next layer up, the spins are antiparallel, and this is the lowest energy configuration for our crystals. However, in our powder sample, the powder data of Chattopadhyay *et al.*,[14] and in the single crystal of Paul *et al.*[9] and Chattopadhyay *et al.*,[15] peaks with integer l are observed. This spin configuration corresponds to the spins in adjacent a-b layers being parallel, rather than antiparallel as shown in Fig. 1. We remark that the dipolar energies for these two configurations are almost identical. Since the dipolar interaction is long range in nature, the energy will be dependent upon defects in the samples such as the density of grain and twin boundaries. We believe that this is the correct explanation for the distinct spin structures which have been observed in both the Gd and Er materials, although considerable work will be needed to establish this connection. We note that a similar duality of spin structures, with identical ordering temperatures, has been observed in other planar antiferromagnets such as Rb_2MnF_4.[13]

Finally, we turn to the temperature dependence of the order parameter as shown in Fig. 3. The left portion shows the data obtained on a single crystal, where the instrumental resolution discriminates against the strong inelastic scattering found along the 2D ridge. The solid curve is a fit of Onsager's exact solution for the $S = \frac{1}{2}$, 2D Ising model,[16] and we see that excellent agreement is obtained. The right portion of the figure shows the equivalent data obtained on the powder sample. The obvious rounding of the data near T_N is due to critical scattering, since in a powder the instrumental resolution necessarily integrates over a substantial fraction of the rod of scattering, along with the Bragg scattering. Thus, powder data more readily exhibit the 2D character of the scattering, but it is difficult to distinguish the inelastic scattering along the rod from the Bragg scattering. Hence, powder data can be quite useful in surveying the overall behavior and revealing the dimensionality of the scattering, but details of the scattering can only be obtained with single crystals of sufficient size.

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